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MEAN FIELD ANALYSIS OF ORIENTATION SELECTIVE GRAIN GROWTH DRIVEN BY INTERFACE-ENERGY ANISOTROPY

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ABSTRACT

Abnormal grain growth is characterized by the lack of a steady state grain size distribution. In extreme cases the size distribution becomes transiently bimodal, with a few grains growing much larger than the average size. This is known as secondary grain growth. In polycrystalline thin films, the surface energy γ_s and film/substrate interfacial energy γ_i vary with grain orientation, providing an orientation-selective driving force that can lead to abnormal grain growth. We employ a mean field analysis that incorporates the effect of interface energy anisotropy to predict the evolution of the grain size/orientation distribution. While abnormal grain growth and texture evolution always result when interface energy anisotropy is present, whether secondary grain growth occurs will depend sensitively on the details of the orientation dependence of γ_i .

INTRODUCTION

The properties of polycrystalline thin films often depend not only on the average grain size, but also on the distribution of grain sizes and on the degree of preferred grain orientation. Orientation selective grain growth in thin films results in an increase of the average grain size with the concurrent development of preferred orientation. This is an *abnormal* grain growth process, i.e., no steady state grain size distribution can result during the transient period in which orientation evolution is occurring. *Secondary* grain growth, an extreme case of abnormal grain growth characterized by the development of a bimodal grain size distribution, is often observed experimentally [1-5] and generally results in a strong preferred orientation. Secondary grain growth can lead to much larger final grain sizes for a given thermal history than would be expected from normal grain growth [6].

Orientation selectivity arises from the presence of crystallographically anisotropic free energies which bias grain growth. The most ubiquitous of these is the anisotropy of the film/substrate interfacial energy and the energy of the film's free surface. We examine here how the functional dependence of the interfacial energy on the relative grain/substrate orientation promotes abnormal grain growth and the development of preferred orientation. A mean field model for grain growth which includes surface and interface energy anisotropy is numerically evaluated to determine the temporal evolution of the grain size/orientation distribution. We show that a bimodal grain size distribution results when the interface energy exhibits a discrete dependence on orientation, but generally does not result for other orientation dependencies. This result suggests that dramatic secondary grain growth, in which grains more than 2 orders of magnitude larger than the surrounding matrix are observed, requires suppression of normal grain growth, e.g., due to grain boundary grooving, in addition to interface energy anisotropy.

MEAN FIELD THEORY AND THE CONNECTION TO GRAIN GROWTH

Hillert [7] first applied the mean field coarsening formalism developed by Lifshitz and Slyozov [8] to grain growth. A variety of modifications of Hillert's approach have since been reported [9].

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Thompson extended the model to include the anisotropy of the surface and interface energy [10]. The physical situation of interest here is shown in Fig. 1. The grains are assumed to be columnar, with film thickness h , and each grain is characterized by its radius R , its surface energy γ_s , and its interface energy γ_i . Both γ_s and γ_i depend on grain orientation, described in general by three angles θ , α , and φ relative to a coordinate system defined for a specific substrate and interface orientation. This results in an orientation dependent mean field growth rate equation:

$$\frac{dR}{dt} = M \left[\frac{\bar{\gamma}_s - \gamma_s}{h} + \frac{\bar{\gamma}_i - \gamma_i}{h} + \gamma_{gb} \left(\frac{1}{\bar{R}} - \frac{1}{R} \right) \right], \quad (1)$$

where $\bar{R}$ and $\bar{\gamma}_{i,s}$ are appropriately defined averages [10]. M is the grain boundary mobility and γ_{gb} is the grain boundary energy. Both are taken here to be constant. The effects of variable M and/or γ_{gb} have been treated using mean field approaches [11,12] and using simulations [13, 14]. This is an alternative source of orientation selectivity which requires that the initial matrix already be highly oriented in order to be observable. In equation (1), grains with lower than the average values of the matrix will tend to have larger growth rates, leading to development of preferred orientation. This effect increases with decreasing film thickness h .

To determine the evolution of the grain size/grain orientation distribution $f(R, \theta, \alpha, \varphi)$, we solve the size flux continuity equation,

$$\frac{\partial f}{\partial t} = - \frac{\partial (f \dot{R})}{\partial R}, \quad (2)$$

where $\dot{R}$ is given by equation (1). Equation (2) is solved numerically in the transient regime, as describe in the next section

Figure 1. Continuous polycrystalline film of thickness h on a substrate.

DESCRIPTION OF THE ANALYSIS

We give only a brief description of the numerical techniques here. More details may be found in [14]. For simplicity we limit the analysis to one orientation degree of freedom such that $\gamma_i = \gamma_i(\theta)$ and $f = f(R, \theta, t)$. This corresponds to a film with all grains having the same crystallographic axis parallel to the substrate surface normal (uniform texture), but having random rotational orientations θ about that axis. The energy of the planar free surface, γ_s , will not vary with rotation about the surface normal and the first term on the right side of equation 1 will be zero. We thus subsequently refer to the interface energy $\gamma_i(\theta)$ simply as $\gamma(\theta)$. Numerical analysis was carried out by computer solution of equation 2 using the Lax-Wendroff finite differencing scheme [15].

We initially examine coarsening under two different functional forms of the interface energy vs. orientation function $\gamma(\theta)$. In the first interface energy function (IEF), $\gamma(\theta)$ varies logarithmically with θ , a common description of low angle boundaries. The second IEF is a step function in γ vs. θ , examined as an alternative approximation to a cusp in $\gamma(\theta)$. These two IEF's are shown in Fig. 2, where we plot the normalized quantity $(\gamma(\theta) - \gamma_0)/\gamma_{gb}$ on the vertical axis and where γ_0 is the average interface energy of the system at $t=0$. The propensity of a system to undergo interface-

energy-driven abnormal grain growth is given by a dimensionless scale factor $Z_0 = \bar{R}_0 \bar{\Delta\gamma}_0^{\max} / h\gamma_{gb}$, where $\bar{R}_0$ is the initial mean particle radius and where $\bar{\Delta\gamma}_0^{\max}$ is defined in Fig. 2. For convenience we introduce the normalized radius $\rho = R/\bar{R}_0$ and the normalized time $\tau = M\gamma_{gb}t/\bar{A}_0$, where $\bar{A}_0$ is the initial mean particle area.

The initial particle size distribution $f(R, \theta, \tau=0)$ input to the program consisted of identical Hillert functions [7], assigned to each of the discrete orientation angles in the range $-\theta_m \leq \theta \leq \theta_m$. The initial particle area per unit angle is constant in all cases discussed here. The particle size distribution, which is what is measured experimentally, is given by $f(R, t) = \sum_j f(R, \theta_j, \tau)$.

Figure 2. A comparison of the step and log interface energy functions (IEF's) used in the analysis.

RESULTS

Figure 3 compares the evolution of the grain size distribution for grain growth under the log and step IEF's with identical normalized driving force Z_0 . It is immediately observed that coarsening under the step IEF produces a transient bimodal particle size distribution (secondary grain growth) while the log IEF results only in abnormal grain growth -- bimodality is not observed. Varying Z_0 by two orders of magnitude (e.g., by varying the film thickness) does not change this observation. Increasing Z_0 does increase the rate of abnormal/secondary grain growth, and, in the case of the step IEF, the degree of bimodality [14].

As abnormal/secondary grain growth occurs, an evolution in the overall crystallographic orientation of the system is also taking place. This is demonstrated in Fig. 4, which shows the grain area at each orientation θ for coarsening under the log and step IEF's. As grain growth progresses, the grain area at the cusp of the IEF's increases preferentially. Coarsening under the log IEF, shown in figure 4a, leads to development of a range of preferred orientations, whereas in Fig. 4b the preferred orientation due to coarsening under the step IEF clearly lies at $\theta < 1^\circ$. The shapes of the curves in Fig. 4 obviously follow directly from the shapes of the input IEF's.

In order to determine whether bimodal grain size distributions occur for IEF's intermediate in shape to the log and step functions, we use an empirically constructed interface energy function:

$$\gamma(\theta) = \frac{a \ln(1 + \frac{60}{\pi} \sin 3\theta) + b[1 - \exp(-\frac{\theta^n}{c})]}{a + b} \quad (3)$$

The first term in the numerator of equation 3 produces essentially logarithmic behavior except that the function obtains zero slope at $\theta_{\max}$. The second term in the numerator produces a simple exponential curve for $n=1$. For $n>2$, the curve becomes s-shaped, approximating a step function. As n is increased, the minimum of the IEF becomes flatter and the transition region between the bottom and top of the curve becomes narrower (more step-like). Decreasing parameter c decreases the width of the cusp. Parameters a and b weight the log-like and step-like contributions.

Since bimodality in the grain size distribution is a transient phenomenon, we need to construct a metric that quantitatively describes the degree of bimodality. Within the context of experimental observations of secondary grain growth, bimodality is large when a few grains grow very large

Figure 3. Time evolution of the grain size distributions (number of grains vs. normalized radius). Left column: coarsening under the log IEF. Right column: coarsening under the step IEF

Figure 4. Time evolution of the area fraction of the film vs. θ . Left column: coarsening under the log IEF. Right column: coarsening under the step IEF.

relative the majority of the grains. Bimodality is small, in this context, when all the grains have similar sizes, or when a few small grains exist within a field of much larger grains. Here we use a bimodality metric that is based on the product of the peak separation times the difference in peak heights. This metric, which satisfies the criteria above and is discussed in ref. [16], is monitored during the numerical analysis. The metric passes through a maximum during abnormal grain growth under any particular IEF and the grain size distribution corresponding to the maximum is output.

In Fig. 5, we vary the shape of the IEF and show the resulting distributions with maximum bimodality metric in each case. In Fig. 5a, we examine the effect of flattening the top of the IEF, i. e., progressively varying the IEF from a condition in which all orientations have different γ , to a condition in which all orientations away from the cusp have the same γ . The latter case is a reasonable description of real IEF's, i. e., orientations away from deep minima have roughly the same γ , with variations that are small compared with the depth of the minima themselves. We see that flattening the top of the IEF produces incipient bimodality, with the extension of a pronounced tail on the distribution to high radius. However, a fully separated secondary peak at large R is not obtained simply by flattening the top of the IEF. In Fig. 5b, we keep the top of the IEF flat, and consider $n=1$, which produces a sharp cusp, and $n=6$, which produces a cusp with a flat bottom

Figure 5. Interface energy functions $\gamma(\theta)$ (left panel of each figure), and the resulting distributions of maximum bimodality (right panel of each figure). (a) Effect of flattening the top of the IEF, with $c=0.7$, $n=1$, and (b) effect of flattening the bottom of the IEF, with $a=0$, $b=1$, $c=0.7$. In (b), only a narrow range of θ near the cusp is shown for clarity.

and a narrower transition region between the bottom and top of the IEF. A true bimodal distribution is observed in the latter case. A more extensive survey, varying parameters a , b , c , and n over a range of values and IEF shapes, results in the same conclusion -- only a step-like IEF produces secondary grain growth and true bimodality of the grain size distribution.

DISCUSSION

We find that the step IEF, a discrete, two-state approximation to the interfacial energy, produces secondary grain growth, characterized by a bimodal grain size distribution and well-defined preferred orientation. The continuously varying log IEF, a more realistic description of interfacial energies, results only in abnormal grain growth, characterized by a monomodal grain size distribution and a range of preferred orientations which evolves with time. Very similar behavior has been observed by Frost et al [18,19], who performed extensive simulations of grain growth using a boundary tracking model that incorporates the essential physical phenomena of grain growth in a direct (but computationally demanding) fashion. Frost *et al* used the step and log IEF's and followed the evolution of the total grain size distribution. They found that grain growth under the step interface energy function resulted in the occurrence of a transient bimodal grain size distribution [19]. However, grain growth under the log IEF with produced only monomodal grain size distributions. These results are in good agreement with our results using the simple mean field approach. From an experimental viewpoint these results are somewhat unexpected, in that *bimodality is frequently observed in real systems undergoing grain growth in the presence of surface or interface energy anisotropy* [1-5]. Inasmuch as all models of interphase boundaries predict smoothly varying energies as a function of orientation near the cusp, we do not believe that observation of secondary grain growth in real systems implies that a step-like IEF is present. Rather, the implication is that, within the context of this model, interface energy anisotropy alone does not produce dramatic secondary grain growth characterized by a highly bimodal size distribution. When secondary grain growth occurs in real systems, it is likely that an IEF such as

that shown in Fig. 5b with $n=1$ is present. However, true bimodality apparently requires additional effects such as grain boundary grooving [17]. Grooving can cause stagnation of the misoriented "normal" grain matrix and promote selective breakaway growth of only a small number of grains with maximum driving force (those that lie at the minimum of the IEF).

CONCLUSIONS

Numerical analysis of a mean field model describing interface energy-driven grain growth in thin films has been presented. It was shown that interface energy anisotropy produces abnormal grain growth, with the concurrent development of preferred orientation as the film coarsens. However, only a step-like IEF produced secondary grain growth, i. e., a bimodal grain size distribution. Since interface energies in real systems are not expected to depend discretely on orientation, this result implies that secondary grain growth follows from a combination of interface energy anisotropy and additional effects such as stagnation of energetically unfavored grains due to boundary grooving.

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